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In this project ~~we have~~ performed both theoretical and experimental studies of optical beam phase-conjugation and electromagnetic scattering processes with intense optical fields. ~~We have established~~ new bounds, on the power and energy required to perform useful wave mixing in certain classes of nonlinear media. ~~We discovered~~ new physical processes, for optical beam phase conjugation. ~~We performed~~ the first complete physical characterizations of monopolar and bipolar photorefractive crystals.

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1. PROJECT OBJECTIVES AND INTRODUCTION

Nonlinear optics, the study of matter interactions with intense electromagnetic fields, has uncovered surprising and useful physical effects, such as stimulated scattering of light by which an intense monochromatic beam can be converted to intense coherent beams of different wavelengths. Recently interesting processes involving nonlinear optical image–bearing beams have been discovered, such as optical–beam phase conjugation. Optical beam phase conjugation is the name given any process which generates, in real time, the time–reversed replica of a complex, image–bearing, optical beam, or which generates other related reflected beams, both monochromatic and polychromatic. Device applications of phase–conjugation include correction of image aberrations, brightness enhancement of laser outputs, and automatic steering of beams, at optical and other wavelengths. Also, such nonlinear optical techniques are being extended to perform many forms of beam processing, sorting, and routing on picosecond time scales. This project aims at exploring and developing these and other new wave–mixing processes, and the necessary nonlinear materials. Other novel electromagnetic scattering processes which occur at the intense optical fields available from lasers are also explored for their scientific and device interest. The approaches taken in this project are both experimental and theoretical.

2. MAJOR ACCOMPLISHMENTS OF THE GRANT PERIOD: 12/03/82 TO 12/02/87

The major accomplishments of this grant were as follows. (1) We constructed the first theoretical study of limiting pump beam requirements for phase conjugation by an optical nonlinearity that is associated with any light scattering process, just as the Kerr effect is associated with Rayleigh scattering. Results promise low power requirements at infrared and longer wavelengths. Experimental efforts to realize the high nonlinearities were begun in this project. (2) We performed the first experimental verification of the prediction of the standard photorefractive model of Kukhtarev for the effects of externally applied electric field on the buildup and decay of photorefractive gratings. We verified that the observed slowing–down of up to twenty–five times neutralized the known enhancement of beam–coupling efficiency. (3) We achieved the first unambiguous measurements of the three impurity parameters needed to predict all photorefractive effects in a simple mono–polar photorefractive material, for changes slower than an electron recombination time. The material used was n–type bismuth silicon oxide. (4) We constructed the first successful theoretical account of electron–hole competition in bipolar photorefractive crystals. (5) We made detailed experimental verification of our theory of electron–hole competition, using bipolar photorefractive bismuth silicon oxide. (6) We performed the first demonstration that phase–conjugation can occur via the well–known two–beam coupling process alone in a photorefractive material with no external or internally generated pump beams. This comprised a totally new physical process for performing optical–beam phase–conjugation. Other accomplishments are noted in the publications and talks listed in the following Sections 3 and 4. More detailed descriptions of our major accomplishments are given in the following subsections 2.1 to 2.6.

2.1. Evaluation of fundamental limits on low power cw phase conjugation at microwave and optical wavelengths.

We have demonstrated theoretically that phase conjugation with reflectivity near unity can be achieved by cw degenerate four–wave mixing in any medium in which beam attenuation arises mainly from scattering with pump beam intensities of order,

$$I_0 = n^2 k_B T \omega^3 / c^2 \tag{1}$$

where n is the refractive index, k_B Boltzmann's constant, T is the absolute temperature, ω is the angular frequency of its beams, and c is the velocity of light. This means pump intensities of only 10^{-4} W/cm^2 will be required for ten micron radiation, or only $\sim 10^{-13}$ W/cm^2 should be required for one centimeter radiation. Similarly lower beam powers will be required to self–focus a coherent beam at longer wavelengths which diffract before being attenuated by scattering alone. This may bear on atmospheric propagation. The theory is described in Publication 3.31. This theory lead to our creation of an experimental program to develop infrared Kerr materials which would realize these high nonlinearities. Preliminary results appear in Publication 3.39.

2.2. Effect of applied electric field on the buildup and decay of photorefractive gratings.

Using a model of simple drift and diffusion for electron transport in the conduction band, Kukhtarev has predicted that, in the more sensitive photorefractive materials, a small applied electric field $E_0(\gtrsim V/\ell)$ will produce: (a) dramatic slowing–down of the writing and erasing of photorefractive charge gratings, and (b)

large temporal oscillations during writing. (Sov. Tech. Phys. Lett. 2, 433, 1976.) Here V = Boltzmann's constant times temperature divided by the electron charge; ℓ = the grating period $\div 2\pi$. This is predicted if ℓ is less than the average distance d moved by an optically excited electron (with $E_0 = 0$) before recombination, but is greater than the Debye screening length. The "hopping" model we developed in this project (Publication 3.8) predicts the same slowing–down, but only for very special probabilities for different hopping distances that are predicted by Kukhtarev. Thus this slowing–down is a critical test of these probabilities. We have made the first quantitative experimental check of this slowing–down, writing a grating of $\ell \sim 0.5$ microns with intersecting 488 nm laser beams in a nominally–undoped crystal $Bi_{12}SiO_{20}$. The above conditions are satisfied. The grating amplitude is monitored by Bragg diffraction of a weak 633 nm laser beam. The internal value of E_0 produced by different applied voltages is determined by the electro–optic effect experienced by the 633 nm beam. We have verified the quantitative predictions of Kukhtarev for the rise and decay functions for 488 nm intensities up to 0.1 W/cm^2 and E_0 up to 8 kV/cm, observing field–induced–slowing of up to 25 times. Details of these results are given in Publication 3.35.

2.3. Optical measurements of the photorefractive parameters of n–type monopolar bismuth silicon oxide.

We achieved the first measurements of all photorefractive parameters of a mono–polar photorefractive crystal necessary to predict all optical interactions, with or without applied electric fields, in the "adiabatic" regime where all process times are long compared to the electron recombination time (~ microseconds). The crystal was n–type bismuth silicon oxide. The parameters are: (a) an effective density of active electron

trap sites (which we find to be 1.4×10^{16} cm^{-3} for 515 nm beams), (b) an average hopping distance or range travelled by an electron in a single excitation–recombination process (which we find to be ~ 3 microns), and (c) the rate of electron excitation per unit light intensity per unit volume (which we find contributes ~ 0.9 cm^{-1} to beam attenuation at 515 nm). This latter measurement with the measured absorption coefficient indicates near unity quantum efficiency. Our measurements were performed using only optical techniques, thus avoiding electrical contact problems inherent in previous methods, which are much more difficult to interpret in any case. Our method was to measure the dependence on writing beam angles and erasure intensities of the decay times of photorefractive holograms. Using the parameter values we determined here, we estimated that an efficient primitive logic operation could be performed with this crystal using as few as 10^5 photons. For details, see Publication 3.32.

2.4. Theory of hole–electron competition in common photorefractive materials.

Recent experiments on some of the most commonly employed photorefractive crystals have indicated that charge transport is not generally of one charge type only, as has been widely assumed in analyzing results with these materials. We have made the first extension of the standard theory of Kukhtarev to treat this competition and verified that it gives an excellent account of previously published photorefractive behavior termed "anomalous". Our theory contains the mobilities, photo–excitation cross–sections, and recombination rate constants of two charge species (instead of one species) in addition to the effective trap density as parameters. Relaxation and beam–coupling data produce curves as a function of grating wavevector which can be critically compared with our predictions, even with the larger parameter space. Details of this work are given in Publication 3.34.

2.5 Experimental verification of the theory of electron–hole competition in a bipolar photorefractive crystal.

We performed the first characterization of both positive and negative charge carriers as well as deep traps in any bipolar photorefractive crystal sufficient to predict photorefractive behavior for all beam geometries on time scales larger than recombination times. By using beam–coupling and light–induced grating–erasure measurements, we found (1) the effective trap density (~ 1ppm), (2) the photorefractive quantum efficiency (near unity), (3) the mobility–lifetime products for electrons and holes, and (4) the excitation cross–sections for electrons and holes in a single crystal of $Bi_{12}SiO_{20}$ of high optical quality. The functional dependencies of measured quantities agreed well with the predictions of our previously–described theory. Details are given in Publication 3.37.

2.6. Optical phase–conjugation by direct backscattering from barium titanate.

We made the first demonstration of a novel process for phase–conjugation in a photorefractive material. This process certainly plays a role in many cases where only four–wave–mixing was previously considered. The new direct backscattering process is an analog of the familiar stimulated Brillouin backscattering, the first nonlinear optical process by which phase–conjugation was achieved. In the photorefractive analog however, there is no frequency shift in the conjugate wave. The threshold for this new process was accurately predicted from previous theory developed in this project (Publication 3.8). Details of experiments and theory are given in Publication 3.33.

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3.30. "Image Processing Using Nonlinear Optical Effects," R.W. Hellwarth, in Optical Nonlinearities, Fast Phenomena and Signal Processing, edited by N. Peyghambarian (National Science Foundation, Washington, D.C., 1986).

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3.39 "Development of colloids of rod shaped β–FeOOH particles in infrared transmitting solvents for use as artificial Kerr media," D.L. Naylor and G. J. Juang (to be published).

4. TALKS, SEMINARS, WORKSHOPS AND CONFERENCES STEMMING FROM THIS PROJECT DURING GRANT PERIOD 12/03/82 TO 12/02/87

4.1. "Optical image processing and computing," Physics Department Seminar, California Institute of Technology, Pasadena, California, 3 May 1983.

4.2. "Band transport model of photorefractive materials with two photoactive levels," G.C. Valley, R.A. Mullen, and R.W. Hellwarth, paper TUM 33, Conference on Lasers and Electro–optics CLEO'83, Baltimore, Maryland, 17 May 1983. Summary published in CLEO'83 Technical Digest, p.74 (Optical Society of America, Washington, D.C., 1983).

4.3. "Wide–angle narrowband optical filter using phase conjugation by stimulated Brillouin scattering," S.K. Saha and R.W. Hellwarth, paper THC4, Conference on Lasers and Electro–optics, Baltimore, Maryland, 19 May 1983. Summary published in CLEO'83 Technical Digest, pp.158–160 (Optical Society of America, Washington, D.C., 1983).

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4.5. "The photorefractive effect for phase conjugation," R.W. Hellwarth, Optics Group Seminar, Royal Signals and Radar Establishment, Gt. Malvern, England, 21 June 1983.

4.6. "Stimulated Brillouin scattering: A collective mode," R.W. Hellwarth, Many–body Seminar, Universite Libre de Bruxelles, Brussels, Belgium, 1 July 1983.

4.7. "Optical beam phase conjugation: A review," R.W. Hellwarth, Physics Seminar, University of Paris VI, 15 July 1983.

4.8. "Sound damping and non–propagating index fluctuations in optical glasses," R.W. Hellwarth, Solid State Seminar, Clarendon Laboratory, Oxford, 24 July 1983.

4.9. "Optical beam phase conjugation: A review," Physics Department, University of New Mexico, Albuquerque, New Mexico, 11 November 1983.

4.10. "Stimulated Brillouin and Raman scattering," R.W. Hellwarth, Hughes Aircraft Company, Electro–optical and Data Systems Group Seminar, 9 December 1983.

4.11. "Non–reciprocal phase shifts from the the photorefractive effect," R.W. Hellwarth, Conference on Physics of Optical Ring Gyros, Snowbird, Utah, 8 January 1984.

4.12. "Error in phase conjugation by stimulated Brillouin scattering," R.W. Hellwarth, 14th Winter Colloquium on Quantum Electronics, Snowbird, Utah, 12 January 1984.

4.13. "Optical beam phase conjugation: A review," R.W. Hellwarth, Institut d'Optique, Orsay, France, 19 April 1984.

4.14. "Phase–conjugation for physical measurement," R.W. Hellwarth, Ecole Normale Superieur, Paris, France, 24 May 1984.

4.15. "The photorefractive effect," R.W. Hellwarth, University of Paris Nord, Paris, France, 15 May 1984.

4.16. "The photorefractive effect: Physics and applications to optical image processing," R.W. Hellwarth, Institut d'Optique, Orsay, France, 17 May 1984.

4.17. "Use of phase conjugation for physical measurement," R.W. Hellwarth, Institut d'Optique, Orsay, France, 29 May 1984.

4.18. "Use of phase conjugation for physical measurement," R.W. Hellwarth, Invited paper ThAA1, XIII International Conference on Quantum Electronics, Anaheim, California, 21 June 1984.

4.19. "Effect of applied electric field on the buildup and decay of photorefractive gratings," J.M. Cohen–Jonathan, R.W. Hellwarth, and G. Roosen, 1984 Annual Meeting of the Optical Society of America, San Diego, California, 31 October 1984, Abstract WBI Technical Program, p.P35.

4.20. "One–way image transmission and reconstruction through a distorting medium," T.Y. Chang and Paul Nachman, paper TuA2, Annual Meeting of the Optical Society of America, 30 October 1984.

4.21. "Phase conjugation, problems and limitations," R.W. Hellwarth, TRW Applied Technology Division Seminar, 10 October, 1984.

4.22. "Physics of optical beam phase conjugation," R.W. Hellwarth, Columbia University Physics Colloquium, 28 September 1984.

4.23. "Physical limits for phase conjugation and self–focusing of electromagnetic waves," R.W. Hellwarth, Aerospace Corporation Seminar, 29 August 1984.

4.24. "Physical limits to optical phase conjugation," R.W. Hellwarth, 15th Winter Colloquium on Quantum Electronics, Snowbird, Utah, 11 January, 1985.

4.25. "Minimum power requirements for efficient four–wave mixing and self–focusing of electromagnetic beams in glasses and fluids," R.W. Hellwarth, Electrical Engineering Colloquium, Ohio State University, Columbus, Ohio, 28 January, 1985.

4.26. "Optical phase conjugation: A review," R.W. Hellwarth, Winter College on Lasers, Atomic and Molecular Physics, International Centre for Theoretical Physics, Trieste, Italy, 10 February, 1985.

4.27. "Phase–conjugation, pulse shortening, and frequency shifting of optical beams by stimulated scattering," R.W. Hellwarth, ibid., 11 February 1985.

4.28. "The photorefractive effect: Physics and applications to optical image processing," R.W. Hellwarth, ibid., 12 February 1985.

4.29. "Use of phase conjugation for physical measurement," R.W. Hellwarth, ibid., 13 February 1985.

4.30. "Minimum energy requirements for optical logic," R.W. Hellwarth, Istituto di Ottica, Florence, Italy, 19 February 1985.

4.31. "Phase conjugation: A review," R.W. Hellwarth, Istituto di Elettronica Quantistica, Florence, Italy, 20 February 1985.

4.32. "Photorefractive measurements," R.W. Hellwarth, Istituto di Ricerce Onde Elettromagnetiche, Florence, Italy, 21 February, 1985.

4.33. "Phase conjugation for four–wave mixing in optical waveguides," R.W. Hellwarth, Physics Colloquium, University of Rome, Rome, Italy, 26 February 1985.

4.34. "Physical limits on logic operations employing nonlinear optical effects," R.W. Hellwarth, paper MC2 at the Winter '85 Topical Meeting on Optical Computing, Incline Village, Nevada, 18 March, 1985. Abstract and Summary in Proceedings.

4.35. "Use of optical phase conjugat'on for understanding basic materials properties," R.W. Hellwarth, Topical Conference on Basic Properties of Optical Materials, National Bureau of Standards, Gaithersburg, Maryland, 9 May 1985. Summary paper published in Basic Properties of Optical Materials, NBS Special Publication 697, edited by A. Feldman (U.S. Government Printing Office, Washington, D.C., 1985).

4.36. "Optical measurements of densities, lifetimes, cross sections and species of charge carriers in insulators and semiconductors," R.W. Hellwarth, Physics Departmental Seminar, Columbia University, New York, New York, 15 November 1985.

4.37. "Fundamental limits on energies and power required for nonlinear optical effects," R.W. Hellwarth, Physics Departmental Seminar, Columbia University, New York, New York, 20 November 1985.

4.38. "Anomalous cross–sections and phase conjugation in optical backscattering," R.W. Hellwarth, Resonance Seminar, Columbia University, New York, New York, 22 November 1985.

4.39. "The photorefraction effect: Physics and application," R.W. Hellwarth, Applied Physics Seminar, Columbia University, New York, New York, 26 November 1985.

4.40. "Optical measurements of electrons and holes in insulators," R.W. Hellwarth, AT&T Bell Laboratories Research Seminar, Murray Hill, New Jersey, 27 November 1985.

4.41. "Determination of electron and hole parameters in insulators," R.W. Hellwarth, Research Seminar, Philips Research Laboratories, Briarcliff Manor, New York, 10 December, 1985.

4.42. "Optical measurements of the properties of photorefractive impurities for device design," R.W. Hellwarth, Invited talk at the Materials Research Society 1985 Annual Meeting, Boston, Massachusetts, 4 December 1985.

4.43. "Photorefractive materials and devices," R.W. Hellwarth, USC Engineering Research Review, Los Angeles, California, 2 April 1985.

4.44. "Fundamental limits on nonlinear optical materials and processes," R.W. Hellwarth, Optical Society of America Workshop on Nonlinear Materials, Annapolis, Maryland, 28 April 1986. Summary in Applied Optics.

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4.48. "Stimulated optical scattering from electron plasma," R.W. Hellwarth, Los Alamos National Laboratory Seminar, Los Alamos, New Mexico, 25 August 1986.

4.49. "Image processing materials," R.W. Hellwarth, Physics Seminar, University of New Mexico, Albuquerque, New Mexico, 28 August 1986.

4.50. "Image processing using nonlinear optical effects," R.W. Hellwarth, NSF Workshop on Optical Nonlinearities, Fast Phenomena, and Signal Processing, University of Arizona, Tucson, Arizona, 21 May 1986. Complete manuscript published in volume of selected papers, Optical Nonlinearities, Fast Phenomena and Signal Processing, edited by N. Peyghambarian (National Science Foundation, Washington, D.C., 1986).

4.51. "Characterization of photorefractive materials," R.W. Hellwarth, Physics Department, University of Amsterdam, Amsterdam, Holland, 1 June 1986.

4.52. "Fundamental limits of power and energy for nonlinear optical effects," R.W. Hellwarth, Institut d'Optique, Orsay, France, 6 June 1986.

4.53. "Optical implementation of cellular automata," R.W. Hellwarth, Physics Department, University of Paris, Place Jussieu, Paris, France, 8 June 1986.

4.54. "Raman–induced phase–conjugation spectroscopy," R.W. Hellwarth, University of Paris, Nord, Paris, France, 13 June 1986.

4.55. "An optical associative memory," R.W. Hellwarth, Max–Planck–Institut für Quantenoptik, Munich, West Germany, 19 June 1986.

4.56. "An optical associative memory," R.W. Hellwarth, Optical Parallel Computing Conference, Utah State University, Logan, Utah, 14 October 1986.

4.57. "Fundamental limits on optical computing," R.W. Hellwarth, Optical Parallel Computing Conference, Utah State University, Logan, Utah, 15 October 1986.

4.58. "Hole–electron competition in photorefractive gratings," F.P. Strohkendl, J.M.C. Jonathan, and R.W. Hellwarth, Paper FQ2 with published abstract. 1986 conference on Lasers and Electro–optics (CLEO), San Francisco, California, 13 June 1986. Abstract in Digest of Technical Papers, pp.386–387, published by the Optical Society of America, Washington, D.C., 1986.

4.59. "Hole contributions to the photorefractive effect in n–type $Bi_{12}SiO_{20}$," F.P. Strohkendl and R.W. Hellwarth, Paper WG17 at the 1986 Annual Meeting of the Optical Society of America; Seattle, Washington, 22 October 1986. Abstract published in the Technical Digest, p.81, published by the Optical Society of America, Washington, D.C., 1986. Short abstract in Optics News, 12, No.9, p.162, September 1986.

4.60. "A comparative study of the photorefractive effect in $Bi_{12}SiO_{20}$ crystals," F.P. Strohkendl, P. Tayebati, and R.W. Hellwarth, at the Topical Meeting on Photorefractive Materials, Effects and Devices, August 12–14, 1987, Los Angeles California. Summary published in Technical Digest Series, 17, pp.32–34, published by the Optical Society of America, Washington, D.C., 1987.

4.61. "Spatial harmonics of photorefractive grating induced by sinusoidal optical intensity variation in a $BaTiO_3$ crystal," Y.H. Lee and R.W. Hellwarth, at the Topical Meeting on Photorefractive Materials, Effects and Devices, August 12–14, 1987, Los Angeles, California. Summary published in Technical Digest Series, 17, pp.64–65, published by the Optical Society of America, Washington, D.C., 1987.

4.62. "Nonlinear refractive–index measurements of air and argon gases at 1 atm," D.M. Pennington, M.A. Henesian, C.D. Swift, and R.W. Hellwarth, 1987 Annual Meeting of the Optical Society of America, 19 October 1987, Rochester, New York. Summary MI6 of Technical Digest, 22, p.28, published by the Optical Society of America, Washington, D.C., 1987.

4.63. "Measurements of anisotropy of parameters in photorefractive barium titanate," T.Y. Chang and R.W. Hellwarth, Conference on Lasers and Electro–optics (CLEO), Baltimore, Maryland, 29 April 1987. Summary published in Technical Digest Series, 14, pp.180–181, published by the Optical Society of America, Washington, D.C., 1987.

4.64. "Stimulated Brillouin scattering in plasmas," R.W. Hellwarth, Los Alamos Scientific Laboratory, Los Alamos, New Mexico, 5 August 1987.

4.65. "Photorefractive Effect in $Bi_{12}SiO_{20}$," F.P. Strohkendl and R.W. Hellwarth, at the Review of Optical Computing and Photonics, University of Southern California, Los Angeles, California, 23 March 1987.

4.66. "Anomalous backscattering from random media," P. Tam and R.W. Hellwarth, at the Review of Optical Computing and Photonics, University of Southern California, Los Angeles, California, 23 March 1987.

4.67. "Cellular automata and optical computing," R.W. Hellwarth, IEEE Student Seminar, University of Southern California, Los Angeles, California, 18 November 1987.

4.68. "Optical devices for neural engineering," R.W. Hellwarth, Center for Neural Engineering (USC) Seminar, University of Southern California, Los Angeles, California, 19 November 1987.

5. PROFESSIONAL PERSONNEL ASSOCIATED WITH THIS PROJECT BETWEEN 12/03/82 and 12/02/87

Principal Investigator:	Robert W. Hellwarth, Professor of Electrical Engineering and Professor of Physics
Postdoctoral Research Associates:	Dr. S.K. Saha Dr. J.M.C. Jonathan Dr. P. Salieri
Doctoral Candidates:	William J. Irons Alessandro Fabretti Chyr–Pwu Tzou Ruth Ann Mullen Tallis Chang Freidrich Strohkendl David Naylor Parviz Tayebati Yeon Ho Lee Patrick Tam Xiaofan Cao

6. ADVANCED DEGREES AWARDED TO PROJECT PARTICIPANTS DURING PERIOD 12/03/82 TO 12/02/87

1. Dr. R. Mullen, Ph.D. Completion: May, 1984. Title of Thesis: "Time–resolved holographic measurements of bulk space–charge gratings in photorefractive $Bi_{12}SiO_{20}$." Current Affiliation: Hughes Research Laboratories, Malibu, California.

2. Dr. T.Y. Chang, Ph.D. completion: December, 1986. Title of Thesis: "Nonlinear optical studies of photorefractive barium titanate: Parameter measurements and phase conjugation." Current Affiliation: Rockwell International Science Center, Thousand Oaks, California.

3. Dr. F. Strohkendl, Ph.D. completion: December, 1987. Title of Thesis: "Photorefractive hole–electron competition, light–induced dark decays, and the photorefractive effect in $Bi_{12}SiO_{20}$. Current Affiliation: University of Southern California, Quantum Electronics Laboratory, Los Angeles, California.

7. INTERACTIONS 12/03/82 TO 12/02/87

The interactions by members of this project are, in addition to the talks, conferences, workshops and seminars listed in Section 4, the consulting services performed by Professor Hellwarth for Lawrence Livermore National Laboratory (contact: Dr. Howard Lowdermilk), Las Alamos National Laboratory (contact: Dr. D.F. Dubois), and the Hughes Research Laboratory (contact: Dr. C.R. Giuliano). The subject matter of this consultation was lasers and laser beam interactions.

8. INVENTIONS

No invention disclosures were filed which stemmed from this effort.